Fractional spinonic excitations in a 1D quantum antiferromagnet $SrCo_2V_2O_8$

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DFG Deutsche Forschungsgemeinschaft

TRR 80 **From Electronic Correlations to Functionality**

EMFL European Magnetic Field Laboratory

Outline

* Experimental techniques: THz/Infrared spectroscopy
  - Quantum spin-dimer system
* 1D spin-1/2 Ising-like XXZ antiferromagnet $SrCo_2V_2O_8$
  - Realization of spinon confinement [PRB **91,** 140404(R) (2015)]
  - Tuning the spinon excitations by external magnetic fields

Spin-1/2 dimer: singlet and triplet

↑ ↓

Cr^{5+} : $3d^1$, S = 1/2

$J_0 \, \mathbf{S}_1 \bullet \mathbf{S}_2$

$J_0 > 0$: antiferromagnetic

Excited state: spin triplet state

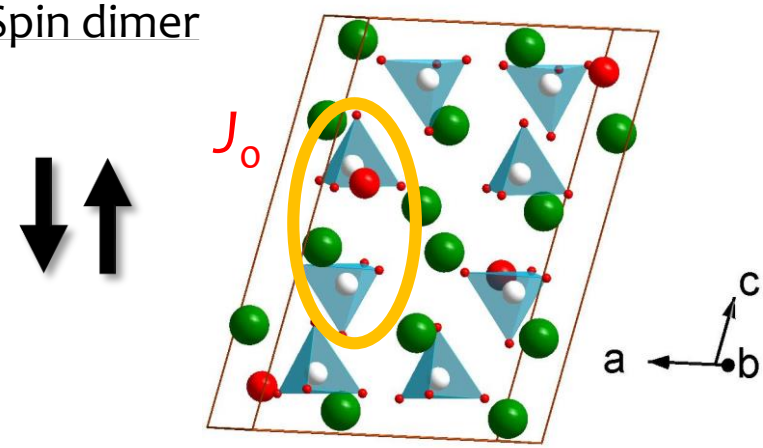
Spin gap $\Delta = J_0$

Ground state: spin singlet state

Singlet-triplet

Monoclinic

Spin dimer



$Ba_3Cr_2O_8$

Zeeman spliting

↑ ↓ Zeeman term

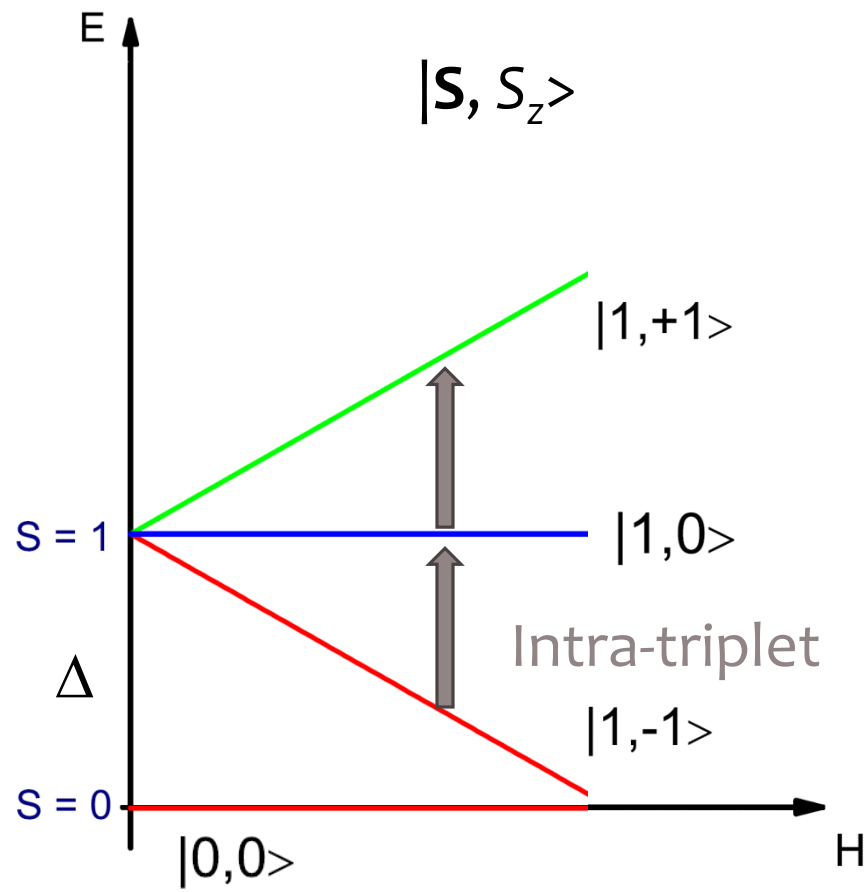
$$J_0 \mathbf{S}_1 \bullet \mathbf{S}_2 + g\mu_B H (S_1 + S_2)^z$$

$J_0 > 0$: antiferromagnetic

Excited state: spin triplet state

Spin gap $\Delta = J_0$

Ground state: spin singlet state



Magnetic excitations

Selection rules:

Intra-triplet mode: $h^{\omega} \perp H$

$$\Delta S = 0, \quad \Delta S_z = \pm 1$$

- Conservation of ***S***
- Magnetic-dipole active

Singlet-triplet mode

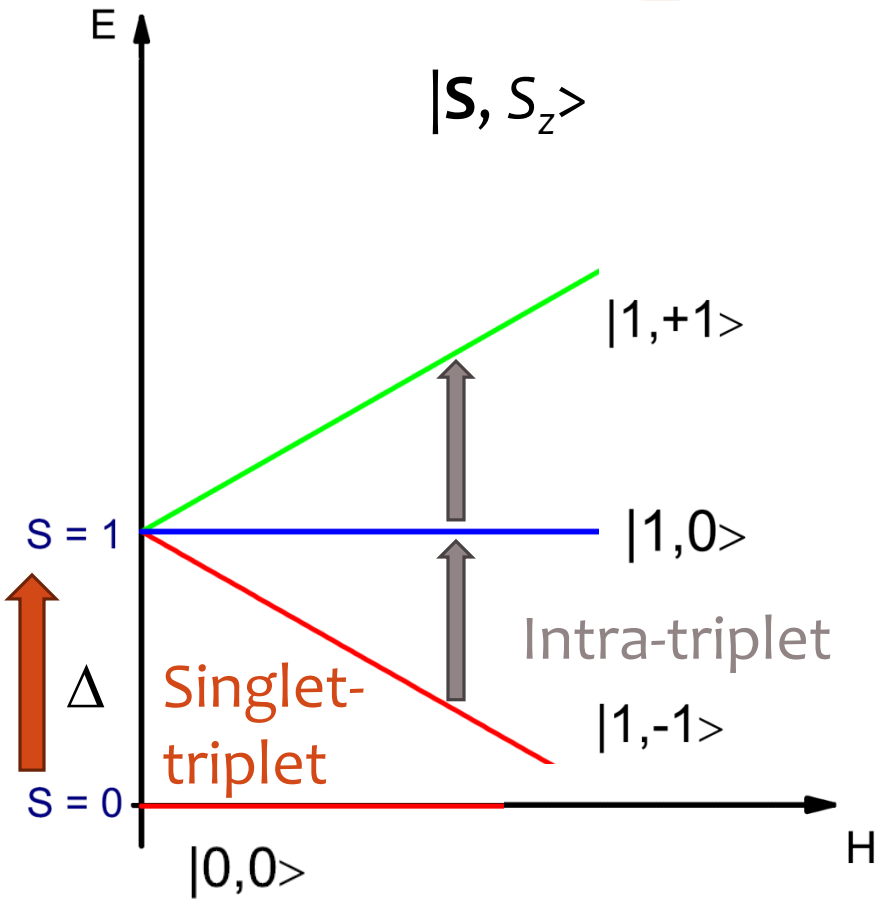
$$\Delta S = 1$$

Violation of *S* conservation!

→ **Dzyaloshinskii-Moriya interaction**

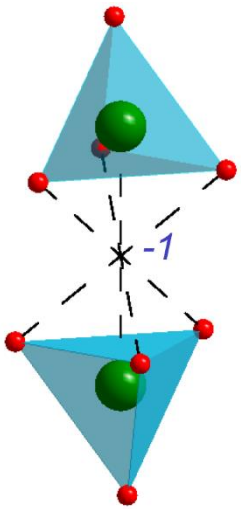
$$\mathbf{D} \cdot (\mathbf{S_1} \times \mathbf{S_2})$$

spin-orbit coupling



Symmetry and selection rule

$Ba_3Cr_2O_8$



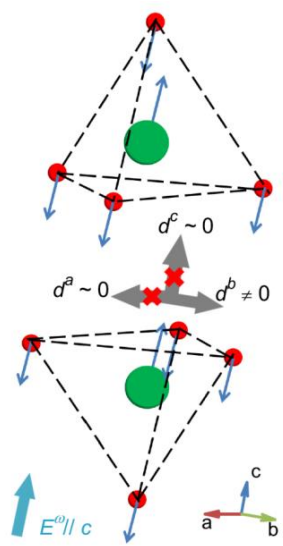
Inversion center

$\mathbf{D} = 0$

T. Moriya, Phys. Rev. **120**, 91 (1960)

Symmetry and selection rule

$Ba_3Cr_2O_8$



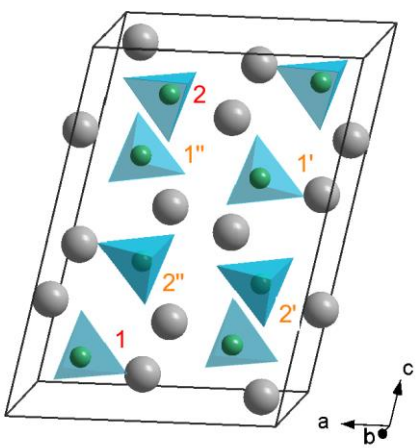
Dynamical Dzyaloshinskii-Moriya: instantaneously symmetry broken due to optical phonons

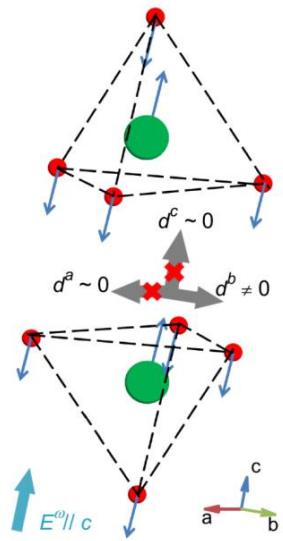
$$\sum_{i,\alpha,\beta} E^{\omega}_{\alpha} A_{\alpha\beta} (\mathbf{S}_{i1} \times \mathbf{S}_{i2})_{\beta}$$

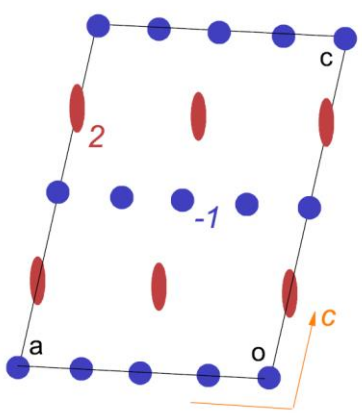
Cépas and Zimmerman, Phys. Rev. B **70**, 024404 (2004)

Symmetry and selection rule

$Ba_3Cr_2O_8$







* Every symmetry operation associates a dimer in one layer to another dimer in the neighboring layer

$$\mathbf{T}_{12} \equiv \mathbf{S}_1 \times \mathbf{S}_2$$

$T^a_{12} \rightarrow T^a_{1'2'}$ q=o in-phase mode

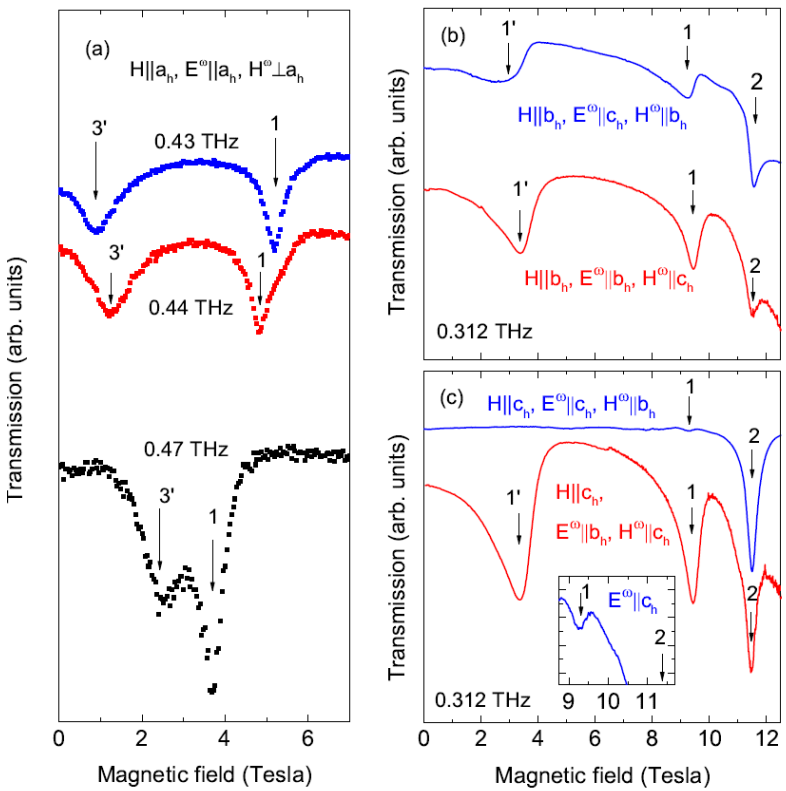
$T^b_{12} \rightarrow -T^b_{1'2'}$ q=π anti-phase mode

Selection rule

$E^\omega \backslash H$	a	b	c
a	$0(A_{ab}) + \pi(A_{ac})$	$\pi(A_{aa}, A_{ac})$	$0(A_{ab}) + \pi(A_{aa})$
b	$0(A_{bc}) + \pi(A_{bb})$	$0(A_{ba}, A_{bc})$	$0(A_{ba}) + \pi(A_{bb})$
c	$0(A_{cb}) + \pi(A_{cc})$	$\pi(A_{ca}, A_{cc})$	$0(A_{cb}) + \pi(A_{ca})$

Experimental results: selection rule and spin-phonon coupling

$Ba_3Cr_2O_8$

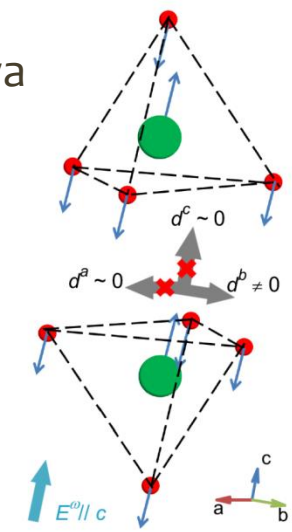


$E^\omega \backslash H$	a_h	b_h	c_h
a_h	$0+\pi$ [Fig. 2(c), 3(b)]		
b_h		$0+\pi$ [Fig. 3(c)]	$0+\pi$ [Fig. 3(d)]
c_h		$0+\pi$ [Fig. 3(c)]	0 [Fig. 2(f), 3(d)]

Dynamical Dzyaloshinskii-Moriya interactions enabled instantaneously due to optical phonons

⬇

spin-phonon coupling



ZW et al, PRB **89**, 174406 (2014)

with O. Cépas, S. Zvyagin

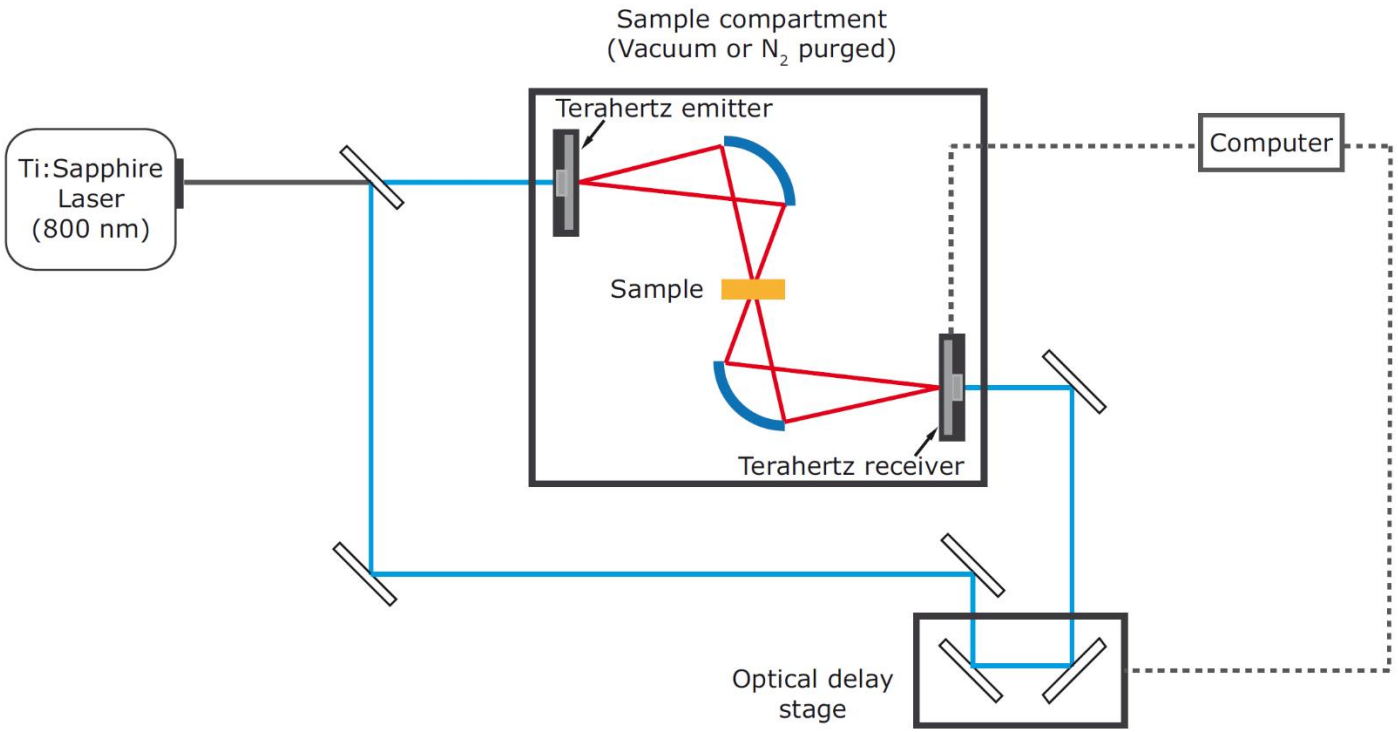
THz and infrared spectroscopy

* Powerful technique for investigating magnetic interactions and dynamics

  characteristic energy range of 1 - 100 meV (THz and FIR spectral range)

* Various tunable parameters:
  - Polarization → selection rules, anisotropies → symmetry
  - Magnetic field → exchange interactions/anisotropies
    → spin hamiltonian
    → classical/quantum phase transitions
    → emergent phenomena
  - Temperature → phase transitions, thermal fluctuations
  - ……

THz time-domain spectroscopy (THz-TDS)

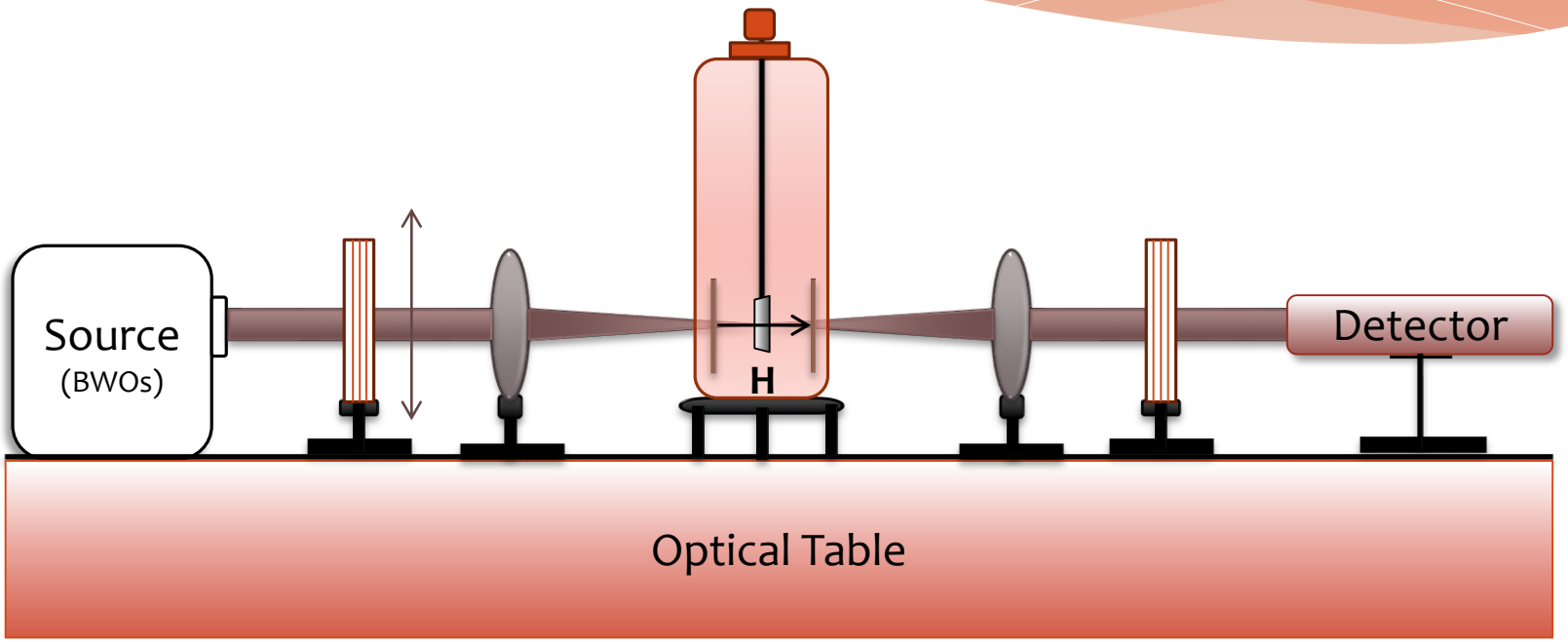


Spectral range:
1 – 16 meV
(0.25 – 4 THz)

Temperature range:
4.2 – 300 K

TeraView Ltd

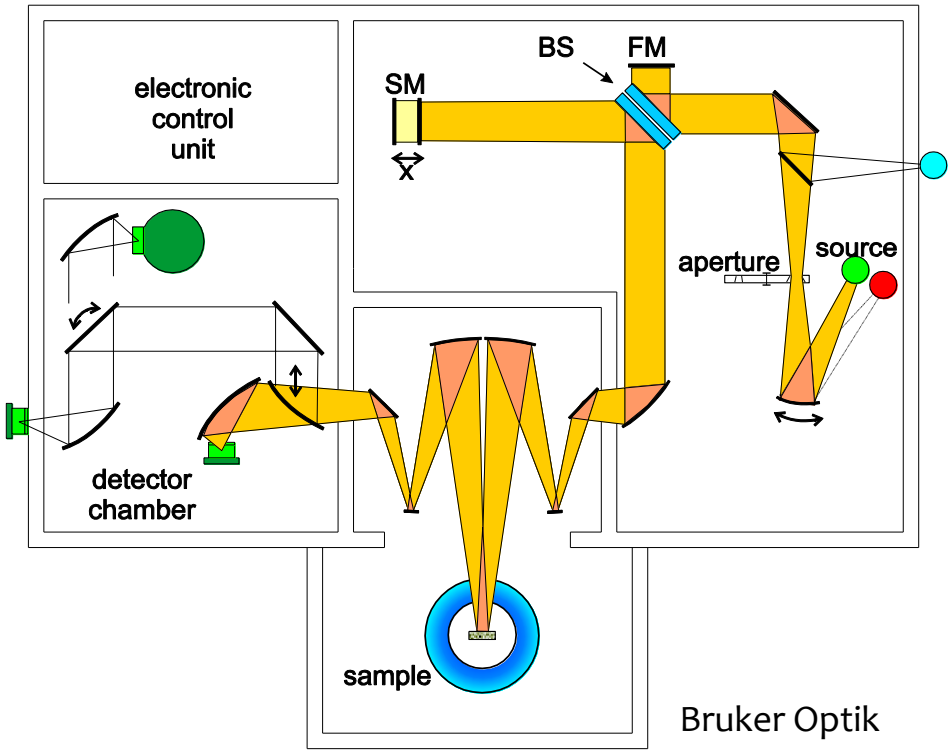
High-field electron spin resonance (ESR) transmission spectroscopy



f	e^i	H	e^o
0.1-1.4 THz (monochromatic)	polarization	7 T 2 – 300 K	polarization

High-field Fourier transform infrared sprectroscopy (FTIR)

Michelson interferometer



Bruker Optik

Bitter magnet (32T)

1.5 – 300 K

© HFML Nijmegen

Frequency domain

Outline

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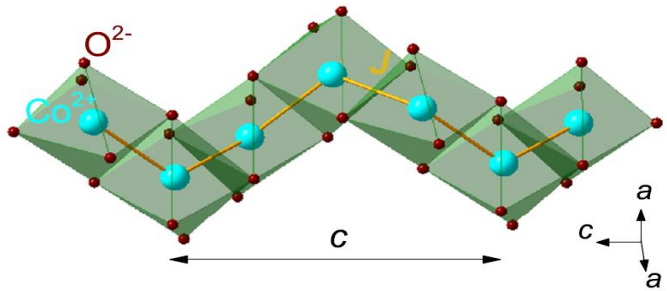
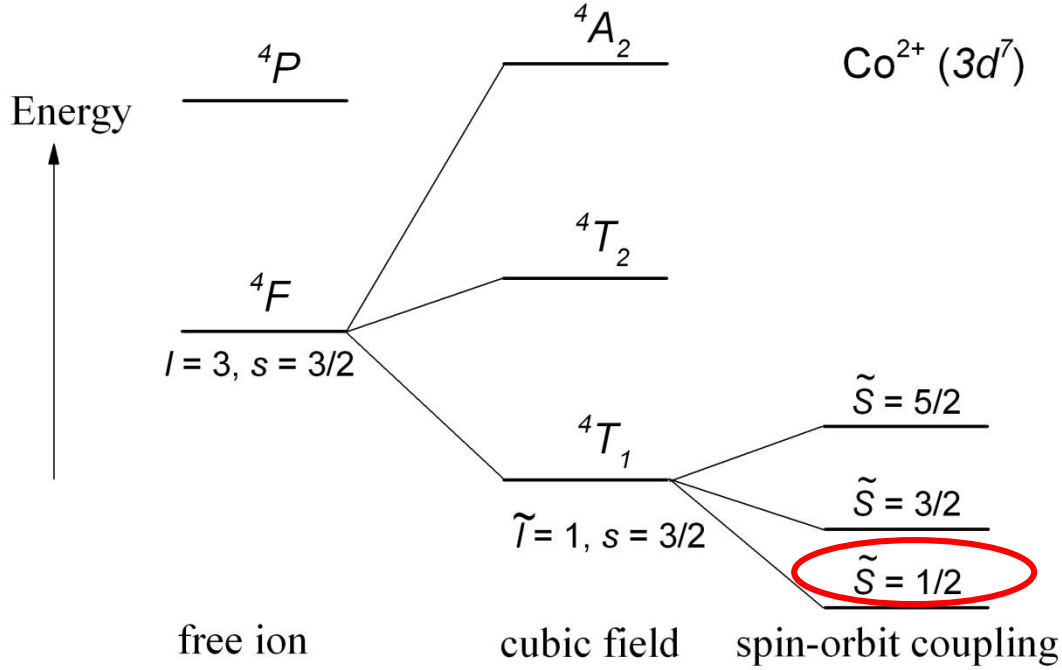
Crystalline and magnetic structure of $SrCo_2V_2O_8$

Tetragonal structure

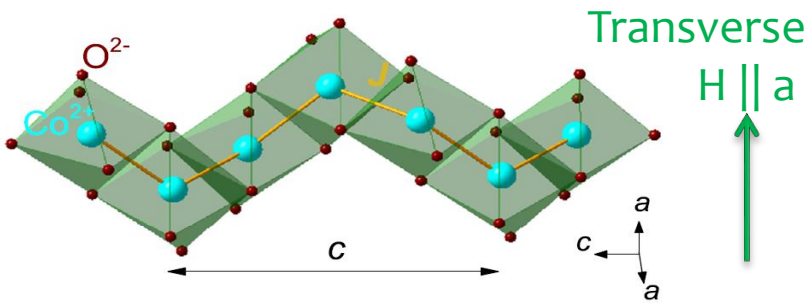




- Screw chain along the c axis
- Edge-sharing CoO_6 octahedra
- 4 Co^{2+} ions per unit cell

Crystalline and magnetic structure of $SrCo_2V_2O_8$

Longitudinal field H || c



Transverse field
H || a

Longitudinal: parallel to the *z* direction (along the *c* axis)

Transverse: perpendicular to the *z* direction (along the *a* axis)

1. Realization of the 1D XXZ spin-1/2 model

$$J\sum_i\left[\boxed{\tilde{S}_i^z\tilde{S}_{i+1}^z}+\boxed{\epsilon(\tilde{S}_i^x\tilde{S}_{i+1}^x+\tilde{S}_i^y\tilde{S}_{i+1}^y)}\right]$$

$J > 0$

antiferromagnetic nearest-neighbor exchange interaction

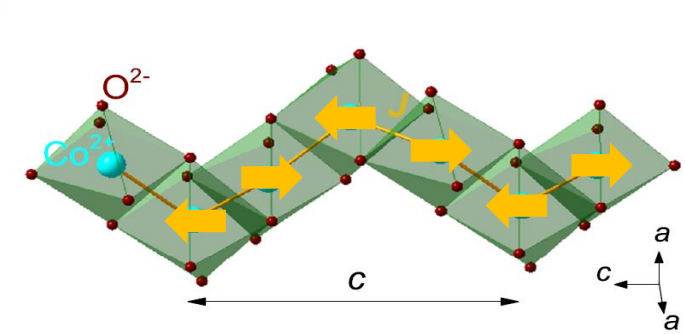
$0 < \epsilon < 1$

Ising-like exchange anisotropy

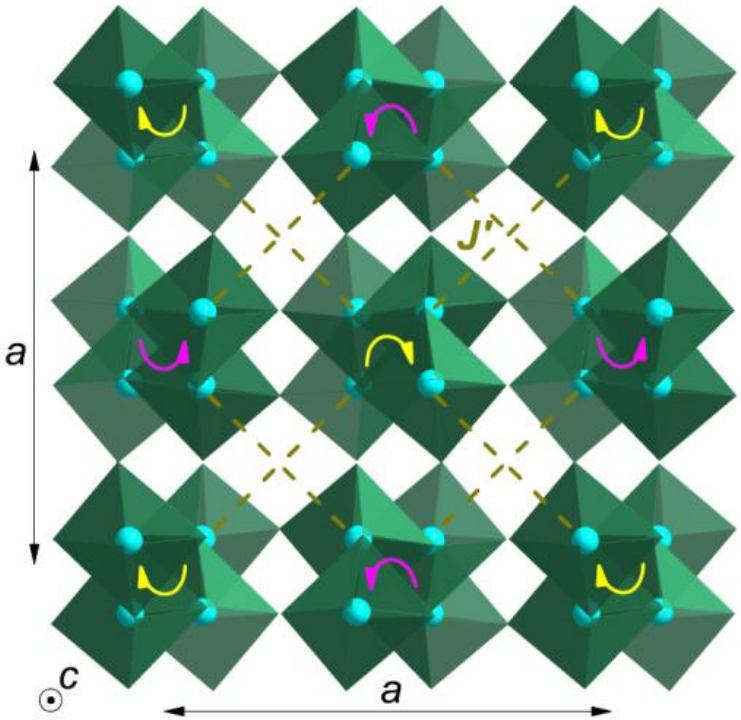
J. C. Bonner and M. E. Fisher, Phys. Rev. **135**, A640 (1964).

Crystalline and magnetic structure of $SrCo_2V_2O_8$

2. Interchain coupling:
   - 3D Néel antiferromagnetic ordering below $T_N \sim 5K$



Neutron diffraction:
A. Bera, B. Lake, et al PRB **89**, 094402 (2014).

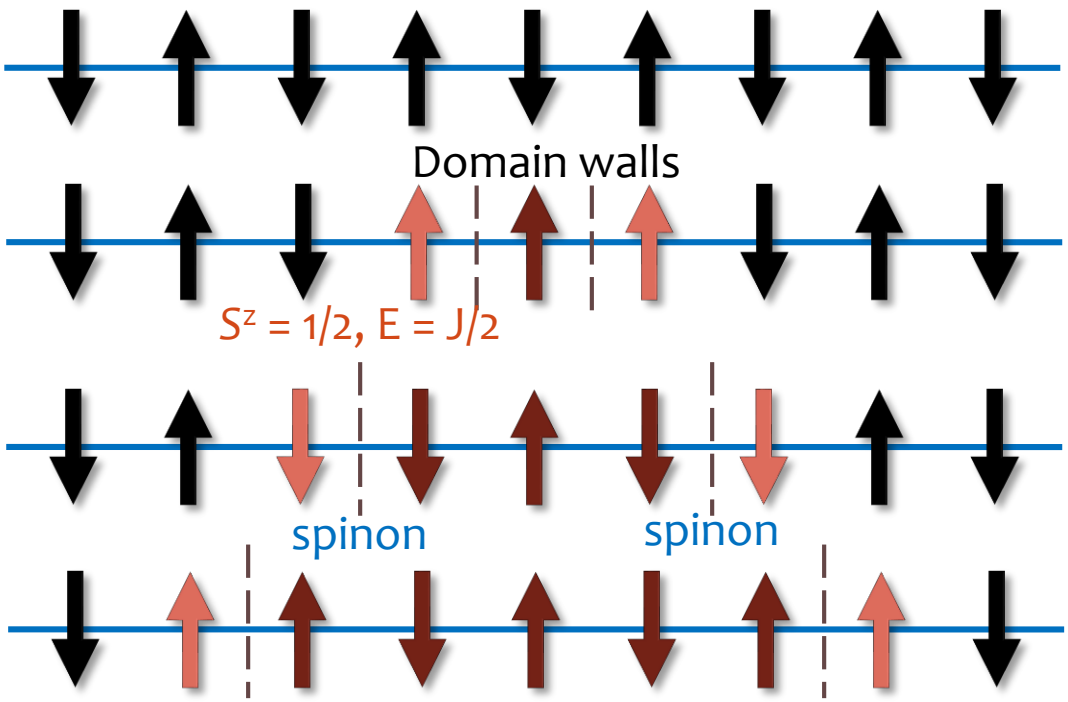


$J'/J < 10^{-2}$

Spinon confinement in 1D Ising-like antiferromagnet

Spinon excitations of Ising chain

Ising antiferromagnetic spin-1/2 chain



$$\sum_{i=1}^{N} J S_i^z S_{i+1}^z$$

One spin flip → Two spinons

$$\Delta S^z = \pm 1$$

$$E = J$$

Highly degenerate excited states

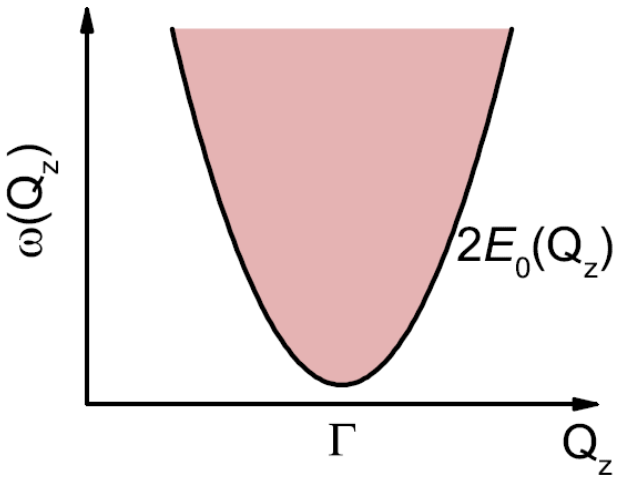
Spinon excitations of XXZ chain

Ising-like (XXZ) spin-1/2 antiferromagnetic chain

$$J\sum_{i=1}^{N}\left[S_i^z S_{i+1}^z + \boxed{\epsilon(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y)}\right]$$

$0 < \epsilon < 1$ (Ising-like)

Excitation continuum



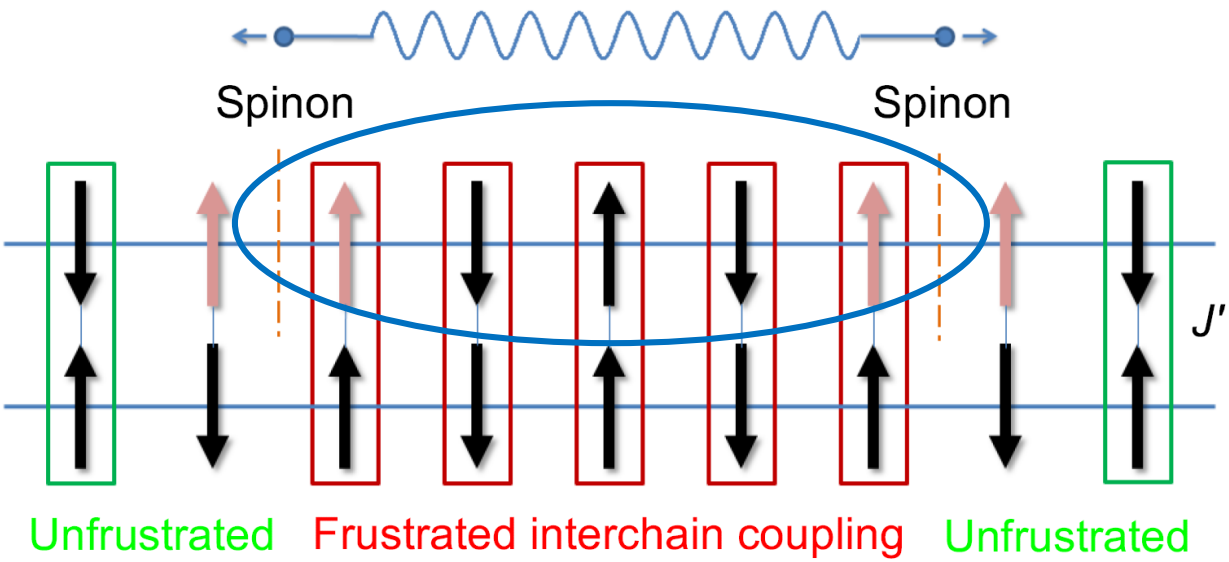
Parabolic dispersion relation
(close to the Γ point, approximate)

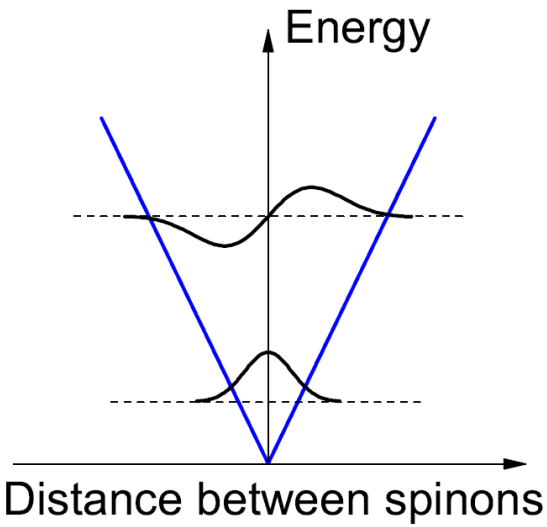
A. H. Bougourzi, et al., Phys. Rev. B **57**, 11429 (1998).

Interchain exchange interaction: confinement potential

$T < T_N$

Two-spinon bound states

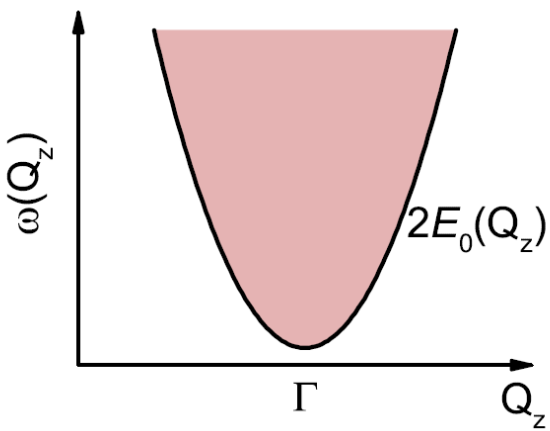


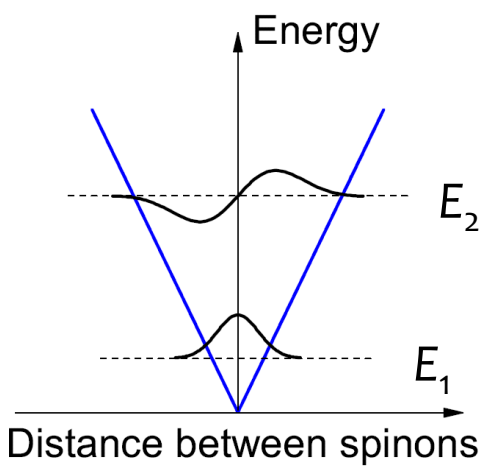


J': interchain coupling

M. Greiter, Nature Physics **6**, 5 (2010).

1D Schrödinger equation of two-spinon bound states





One-dimensional Schrödinger equation:

$$-\frac{\hbar^2}{\mu}\frac{d^2\varphi}{dz^2} + \lambda|z|\varphi = (E - 2E_0)\varphi$$

Solution:

$$E_j = 2E_0 + \zeta_j \lambda^{2/3}\left(\frac{\hbar^2}{\mu}\right)^{1/3}$$

ζ_j are the negative zeros of the Airy function

Linear dependence:
Characteristics of confined spinons

B. M. McCoy and T. T. Wu, PRD **18**, 1259 (1978).

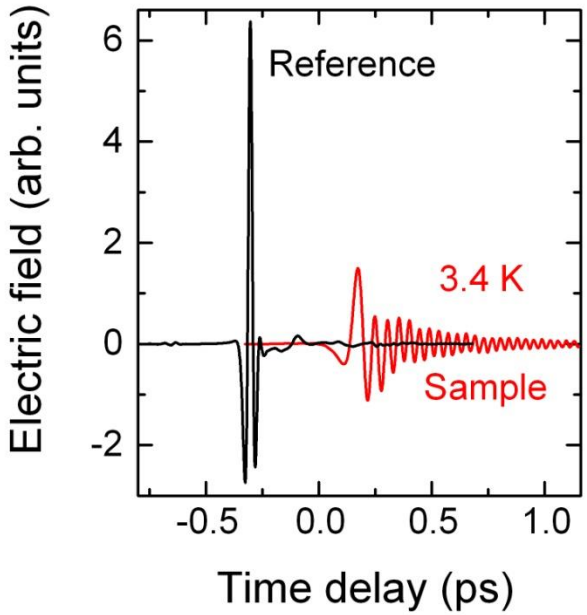
Result I: Realization of spinon confinement

Spinon absorption spectra

$SrCo_2V_2O_8$

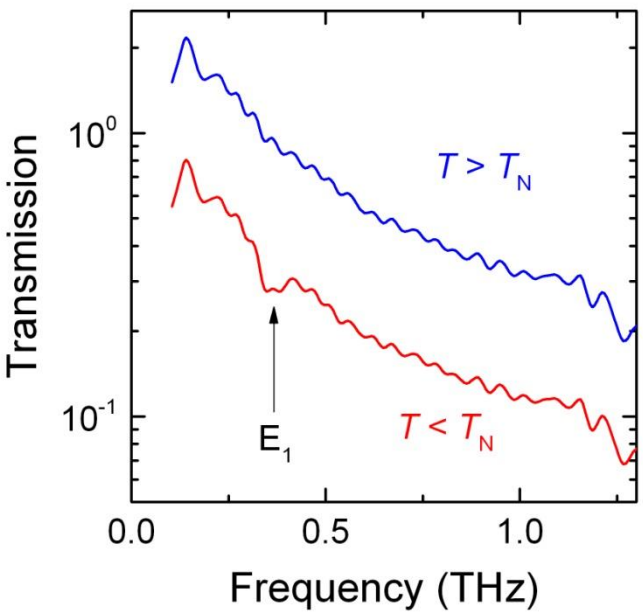
$T_N \sim 5K$

THz Time domain signal



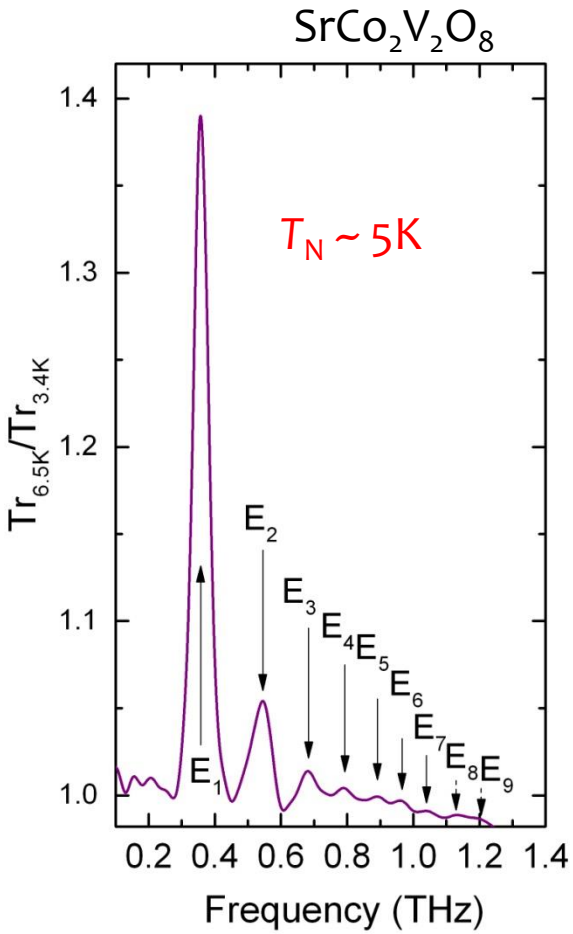
Fourier Transform

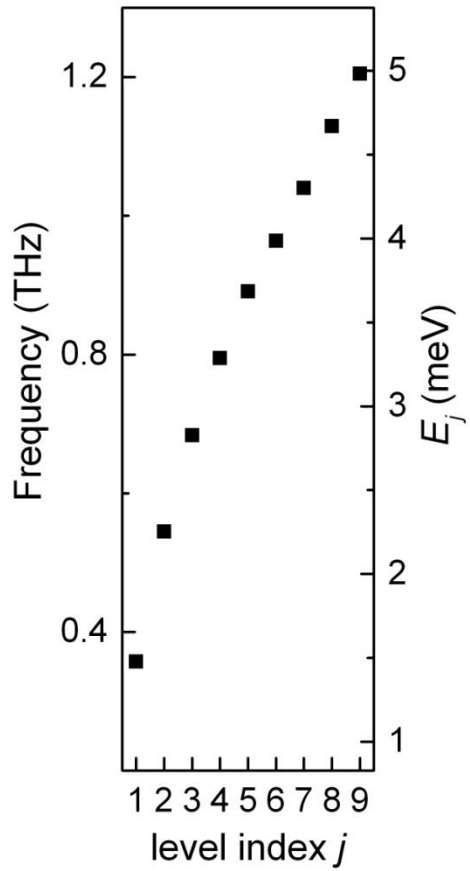
Frequency domain signal



ZW et al PRB **91,** 140404(R) (2015)

Spinon absorption spectra





Series of excitations:

- Below T_N : magnetic order

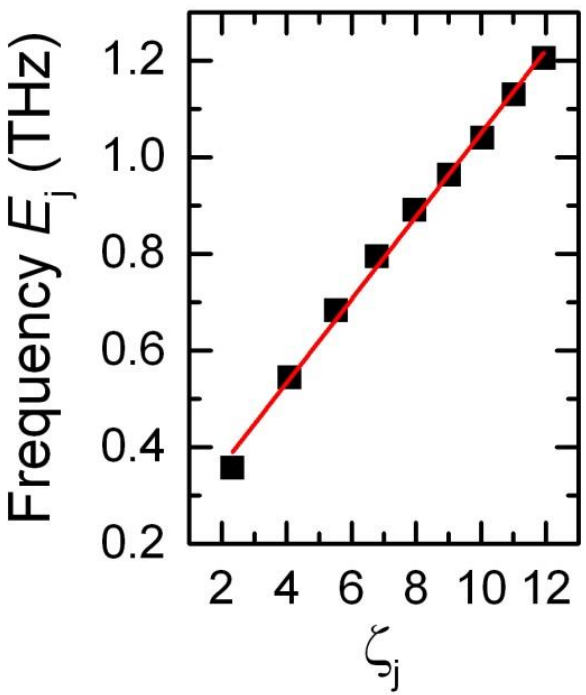
Characteristic features:

- Energy increases
- Intensity decreases
- Energy difference decreases

ZW et al PRB **91,** 140404(R) (2015)

Series of confined spinons

$SrCo_2V_2O_8$



$2E_0$ = 0.96 meV

$\lambda^{2/3} (\hbar^2/\mu)^{1/3}$ = 0.33 meV

One-dimensional Schrödinger equation with linear confinement potential:

$$-\frac{\hbar^2}{\mu}\frac{d^2\varphi}{dz^2} + \lambda|z|\varphi = (E - 2E_0)\varphi$$

Solution:

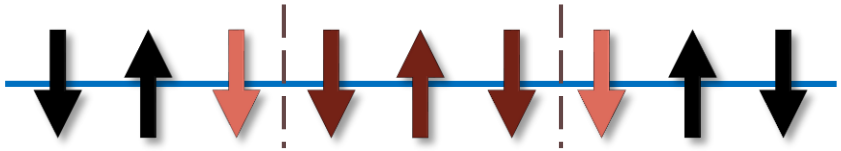
$$E_j = 2E_0 + \zeta_j \lambda^{2/3}\left(\frac{\hbar^2}{\mu}\right)^{1/3}$$

ζ_j are the negative zeros of the Airy function

B. M. McCoy and T. T. Wu, PRD **18**, 1259 (1978).

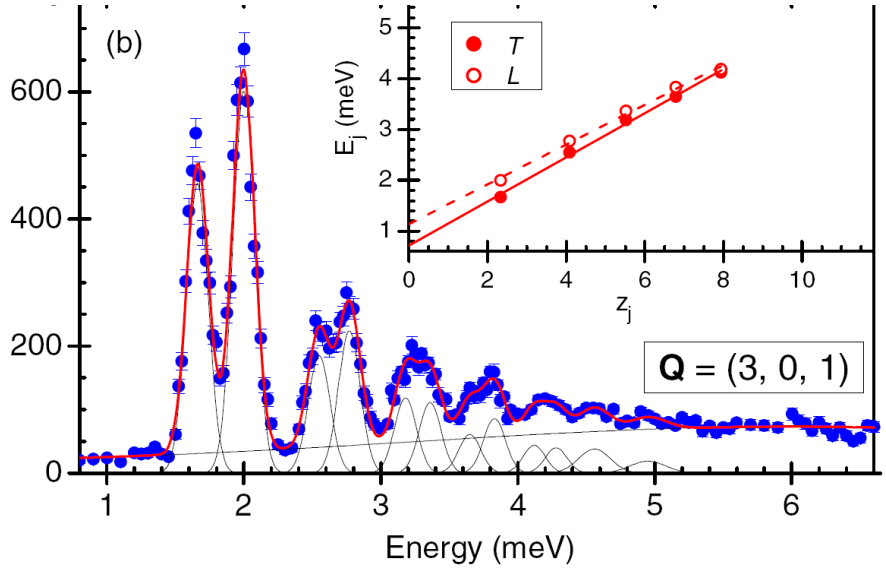
Discussion: selection rule

- Two-spinon bound state S =±1:
  odd number of spin flips



- Two-spinon bound state S =0:
  even number of spin flips

$BaCo_2V_2O_8$



both series observed by neutron scattering

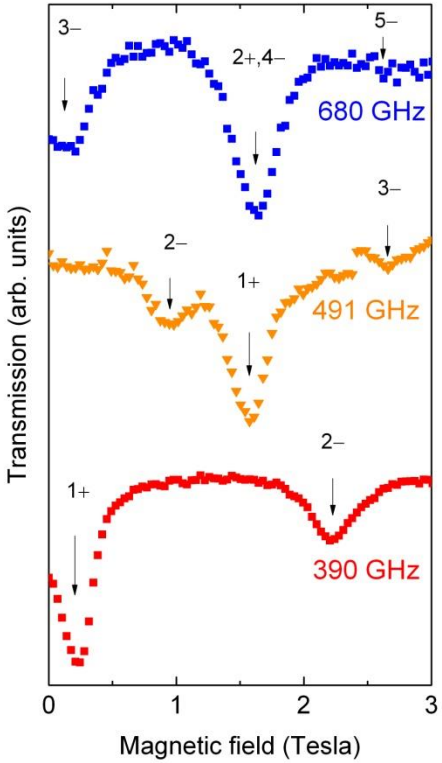
B. Grenier et al PRL (2015)

Result II: magnetic-field tuning

Longitudinal magnetic field

… in a *longitudinal* magnetic field

High-field ESR $H \parallel z$

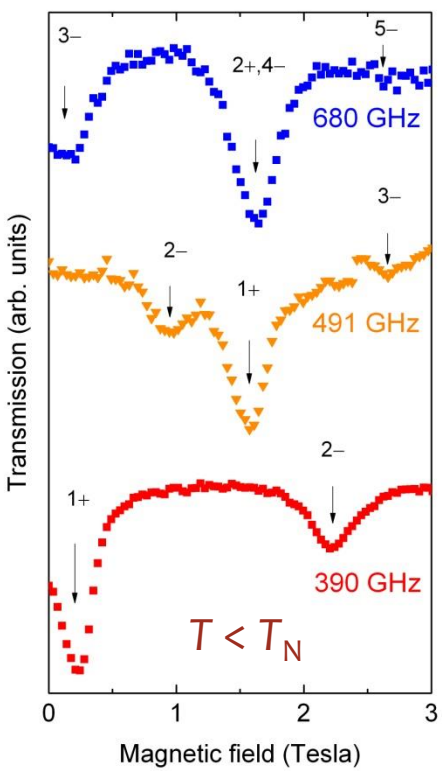


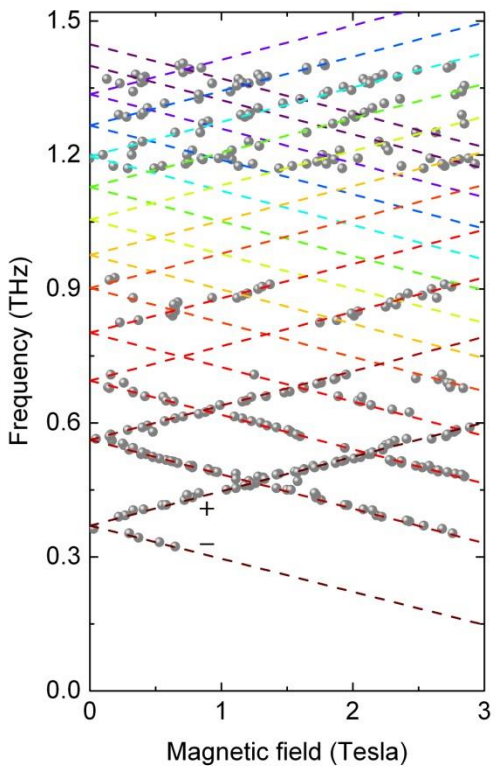
$SrCo_2V_2O_8$ $T < T_N$

ZW et al PRB **91,** 140404(R) (2015)

… in a *longitudinal* magnetic field

High-field ESR $H \parallel z$





Linear field dependence described by a single Zeeman term

$$\pm g_{\parallel} \mu_B H S$$

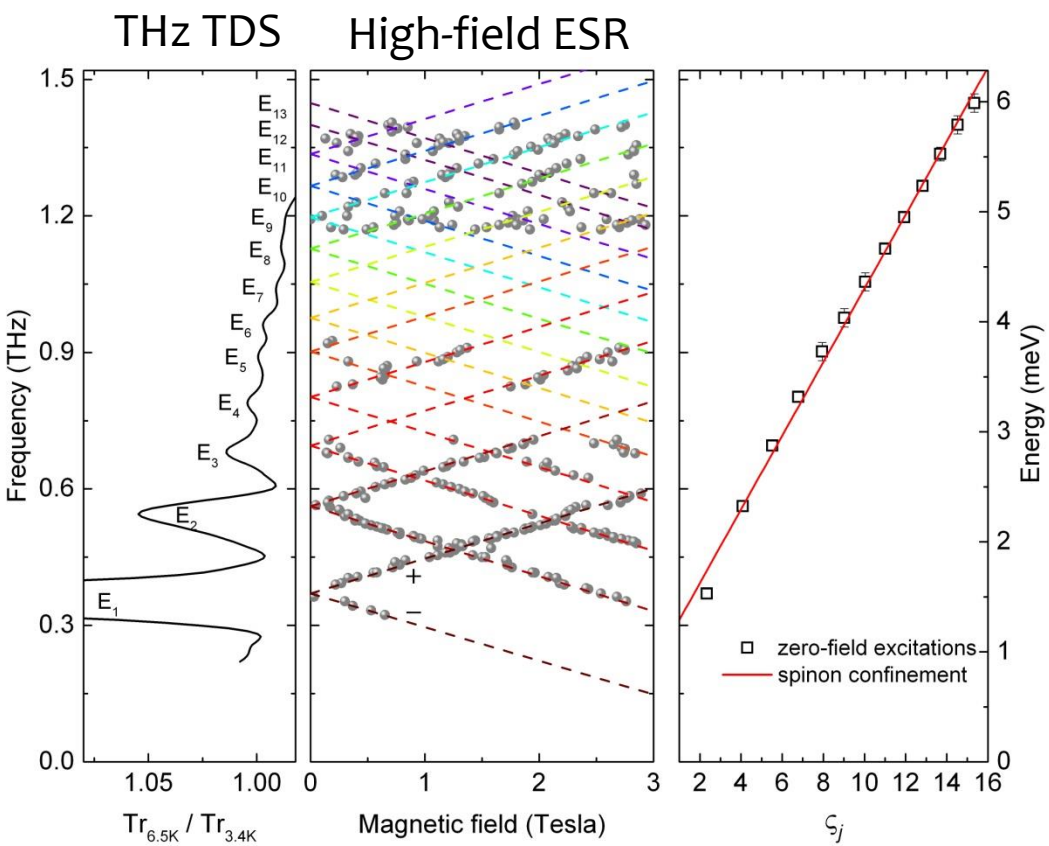
$$g_{\parallel} = 5.5 \qquad S = 1$$

(Distortion of CoO_6 octahedra)

ZW et al PRB **91,** 140404(R) (2015)

Split of bound states in longitudinal magnetic field

$H \parallel z$



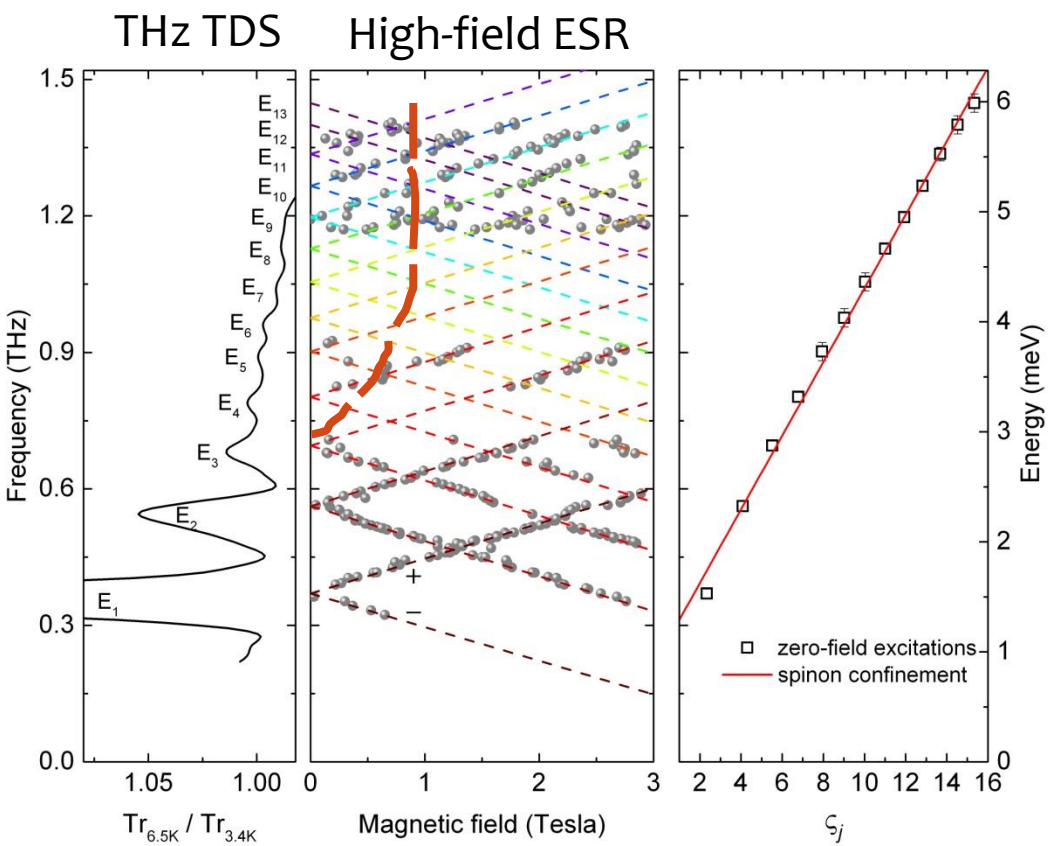
$$E_j = 2E_0 + \zeta_j \lambda^{2/3} \left(\frac{\hbar^2}{\mu}\right)^{1/3}$$

- Higher-energy confined spinons determined from linear extrapolation of field dependence
- Two-spinon bound state doubly degenerate at zero field and split in finite longitudinal field
- Two-spinon bound state S =±1

ZW et al PRB **91,** 140404(R) (2015)

4-spinon continuum, excitations of the critical spin-liquid phase, etc

H || *z*



- Two pairs of bound-spinons *continuum* to be checked (e.g. linewidth)?
- Excitations in the critical region?
- Field dependence in $BaCo_2V_2O_8$

Result II: magnetic-field tuning

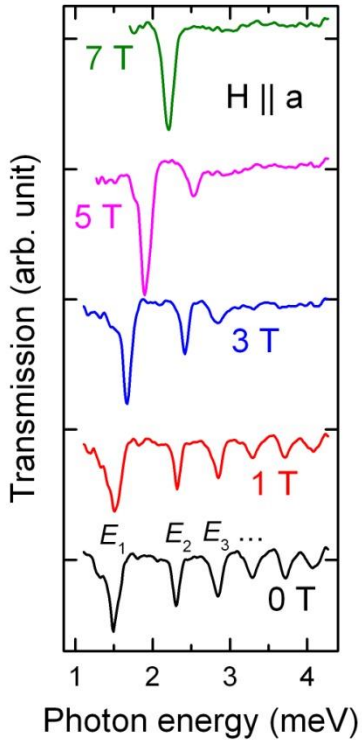
Transverse magnetic field

in the antiferromagnetic phase

$H \parallel x$

$T < T_N$

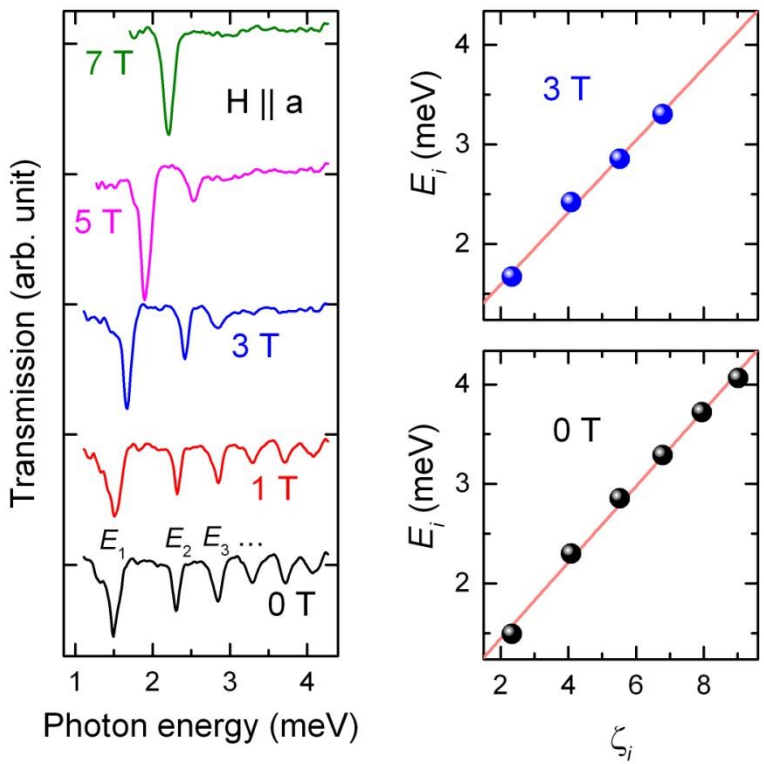
High field FTIR



ZW et al, unpublished

in the antiferromagnetic phase

High field FTIR



ZW et al, unpublished

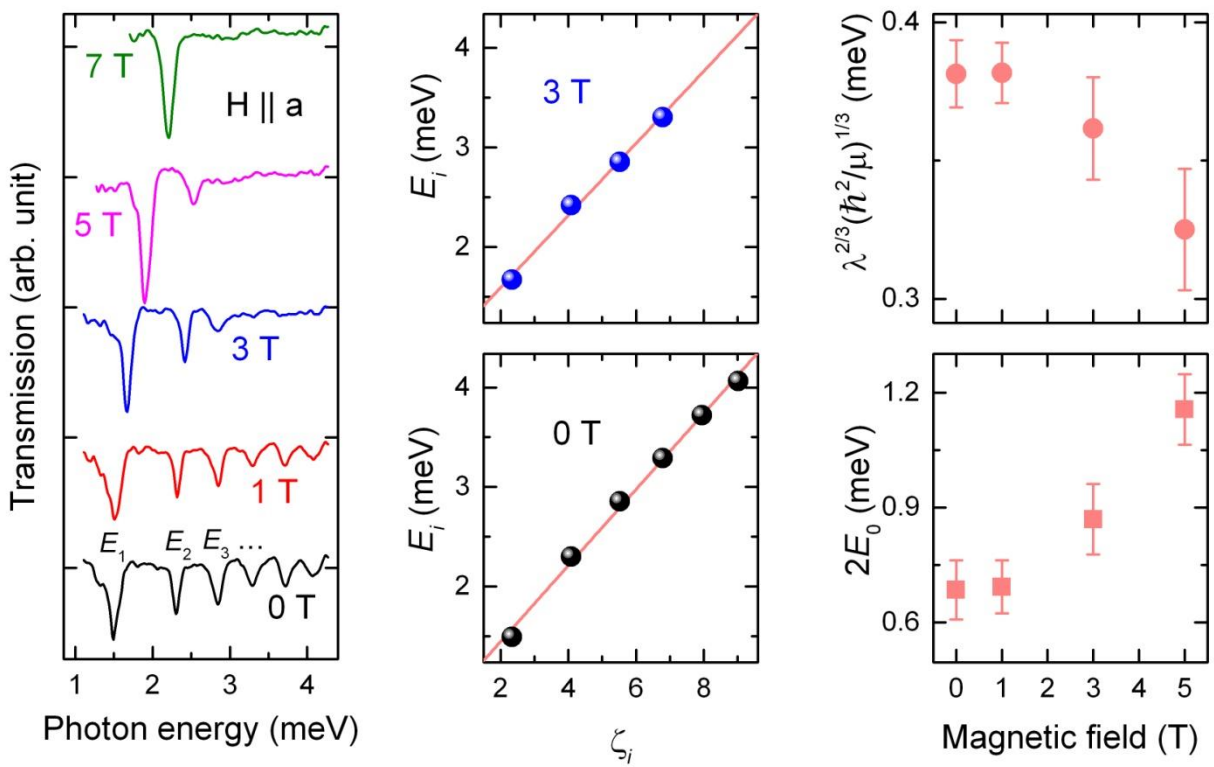
One-dimensional Schrödinger equation with linear confinement potential:

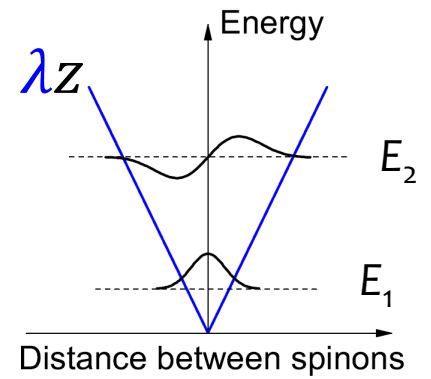
$$-\frac{\hbar^2}{\mu}\frac{d^2\varphi}{dz^2} + \lambda|z|\varphi = (E - 2E_0)\varphi$$

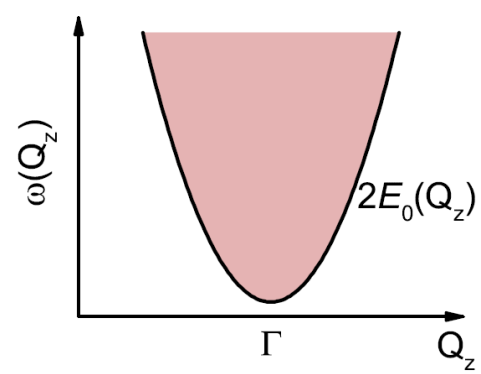
$$E_j = 2E_0 + \zeta_j \lambda^{2/3}\left(\frac{\hbar^2}{\mu}\right)^{1/3}$$

in the antiferromagnetic phase

$$E_j = 2E_0 + \zeta_j \lambda^{2/3} \left(\frac{\hbar^2}{\mu}\right)^{1/3}$$



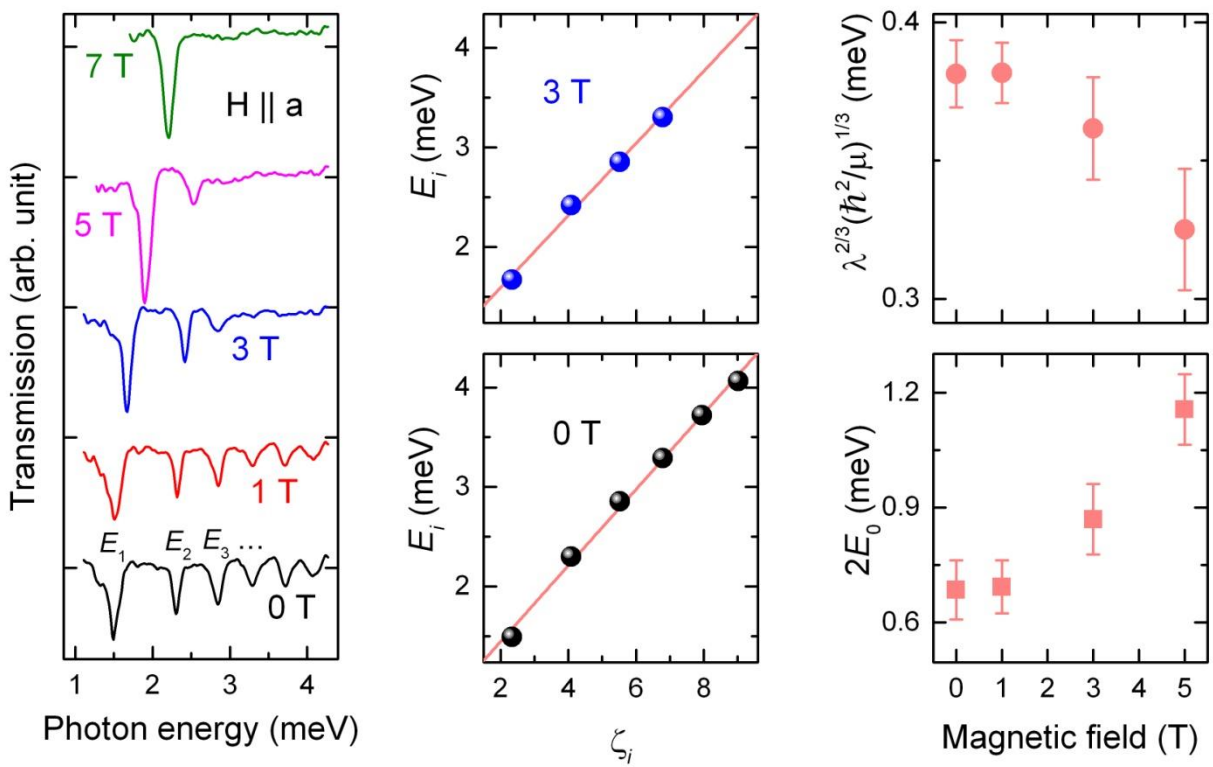


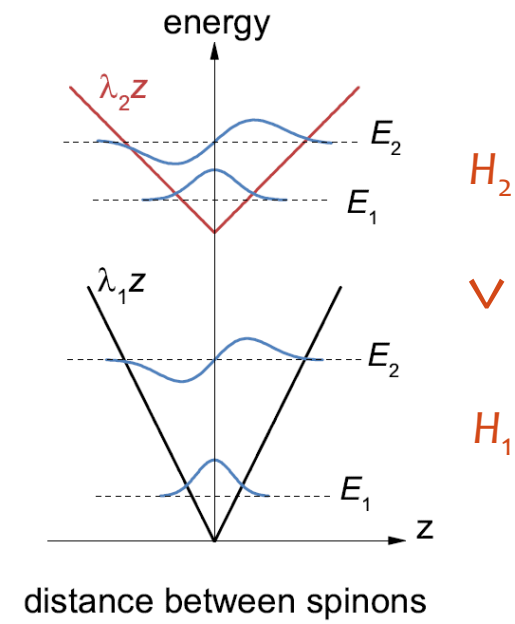


ZW et al, unpublished

in the antiferromagnetic phase

$$E_j = 2E_0 + \zeta_j \lambda^{2/3} \left(\frac{\hbar^2}{\mu}\right)^{1/3}$$

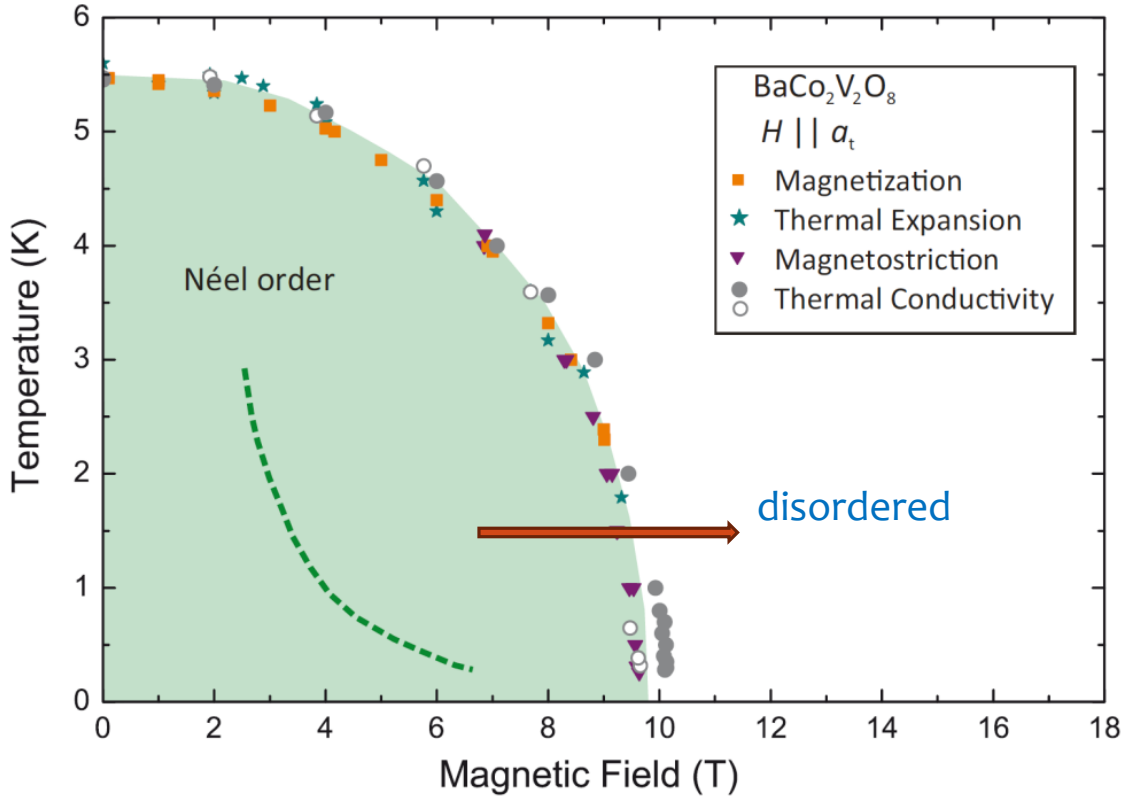




field-induced transition:
$H_c \sim 7$ T

ZW et al, unpublished

Low temperature phase diagram of $BaCo_2V_2O_8$ in transverse magnetic fields



$BaCo_2V_2O_8$:
critical field
$H_c \sim 9$ T
at 1.5K

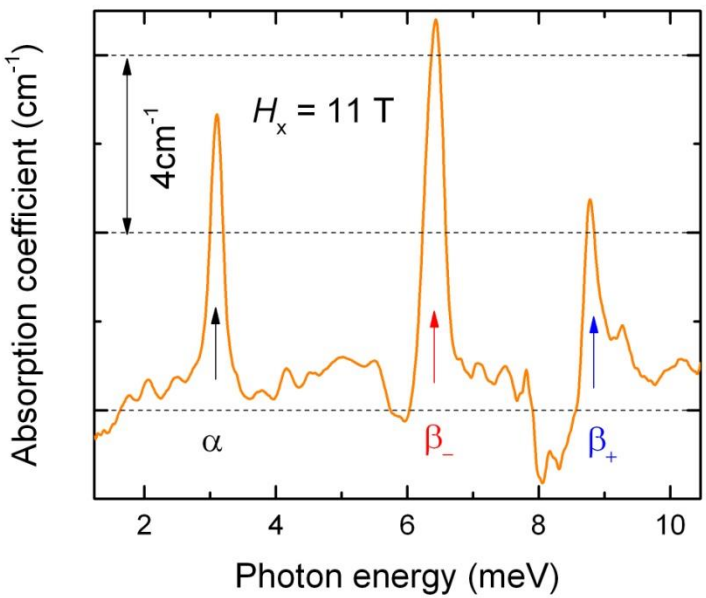
$SrCo_2V_2O_8$:
$H_c \sim 7$ T
at 1.5K

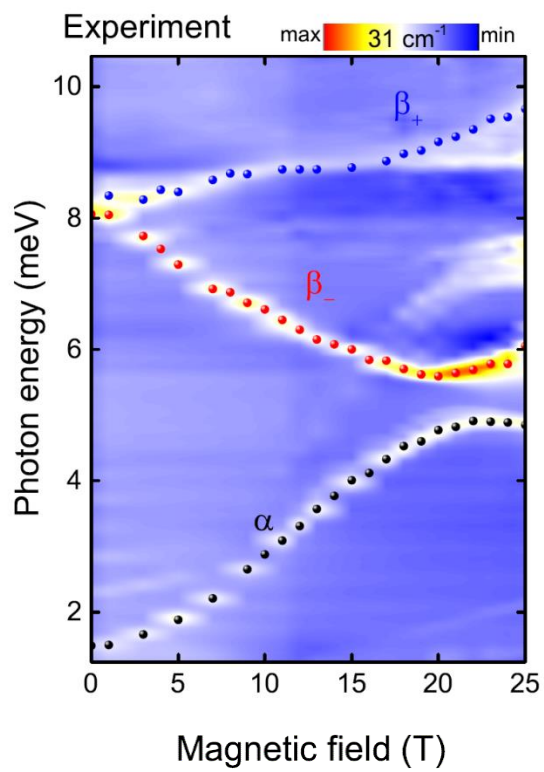
S. K. Niesen, Th. Lorenz *et al*. PRB **87**, 224413 (2013)

in the disordered phase

$H \parallel x$

High field FTIR





Field dependence:

- α evolves from confined spinons
- β_+, β_- are degenerate at zero field and split in finite field
- α, β_- avoid crossing at high fields above 22T

ZW et al, unpublished

Summary

* Observation of confined spinon excitations in the 1D XXZ antiferromagnet $SrCo_2V_2O_8$
  → linear confinement potential by interchain coupling
  → described by one-dimensional Schrödinger equation
* Tuning of spinon excitations by magnetic fields
  Longitudinal magnetic field
  → linear dependence in the ordered phase
  Transverse magnetic field
  → non-linear field dependence